THE COLLISIONAL EXCITATION OF NITROGEN BY LOW ENERGY ELECTRONS TO THE SECOND POSITIVE ($C^3\Pi_u$) TRANSITION

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THESIS

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by

Steven Wayne Ader
and
William Charles Doerner

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When the nitrogen was bombarded by 250 eV electrons the effective cross section was determined to be $(1.72 \pm .22) \times 10^{-17}$

Continued from block #20

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The Collisional Excitation of Nitrogen by Low Energy Electrons to the Second Positive ($C^3\Pi_u$) Transition

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When the nitrogen was bombarded by 250 eV electrons the effective cross section was determined to be $(1.72 \pm .22) \times 10^{-17}$ cm^2, which was the maximum value of the cross section measured.

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I. INTRODUCTION

The measurement of effective excitation cross-sections for molecular nitrogen is important in the understanding of upper atmosphere phenomena such as aurora and afterglow.

The excitation of nitrogen to the $C^3\Pi_u$ by electrons is as follows:

$$e^- + N_2\ (X^1\Sigma_g^+) \rightarrow N_2\ (C^3\Pi_u) + e^-.$$

The second positive transition ($C^3\Pi_u \rightarrow B^3\Pi_g$) follows and light is emitted from the lowest vibrational level in the v" progression (0,0), (0,1), (0,2), (0,3), (0.4), (0.5). The transition probabilities for these transitions is well known [Ref. 3]. The transition of highest probability is the (0,0) transition at a wavelength of 337.13 nm.

The excitation cross section for this transition was measured for bombardment by electrons in the energy range 80 to 900 electron volts. It had been hoped that measurements could be made at lower energies but this was precluded by equipment difficulties which will be explained later.

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II. THE CROSS SECTION EQUATION

A. THEORY

In discussing the excitation of the molecule of interest, we may write for the rate of change of state K

$$(1) \quad \frac{dN_K^*}{dt} = R_K - \sum_{f=0}^{\infty} \lambda_{0f} N_K^* - Q$$

where N_K^* is the population of the excited state K, R_K is the rate of populating the excited state K by electron-molecule collisions, λ_{0f} is the relative transition probability to a final state f, and Q is the collisional deexcitation rate.

If $\bar{t}$ is the mean life of the excited state K and t the time of electron bombardment, $t \gg \bar{t}$ implies that a steady state situation exists among the populations of the various excited and final states. Therefore, we may write

$$(2) \quad R_K = \sum_{f=0}^{\infty} \lambda_{0f} N_K^* + Q.$$

If the gas pressure is sufficiently low so that the mean time between collisions $\bar{t}_c$ is small compared with the mean life of the excited state $\bar{t}$, then Q, the collisional de-excitation rate, may be ignored in comparison with the other terms. The mean time between collisions is

$$\bar{t}_c = \frac{\bar{l}}{v}$$

where $\bar{l}$ is the mean free path of the molecules and v is the molecular thermal velocity. If we assume the nitrogen gas

may be reasonably approximated as being ideal, we obtain

$$\bar{l} = \frac{1}{\sqrt{2}\ \pi d^2 n}$$

where d is the molecular diameter, and

$$v = \frac{\sqrt{3kT}}{\sqrt{m}}$$

with m the molecular mass. This leads to

$$\bar{t}_c = \frac{\sqrt{mkT}}{\sqrt{6}\ \pi d^2 P} \quad .$$

For the (0,0) transition, $\bar{t} = 10^{-7}$ seconds [Ref. 3]. If we choose $\bar{t}_c = 10^3 \bar{t}$ as sufficiently large for our approximation to be valid, we obtain $P \lesssim 3 \times 10^{-3}$ torr as an upper limit for eliminating Q from consideration.

As a result, we may now write (for excitation at low pressures)

$$(3) \qquad R_K = \sum_{f=0}^{\infty} \lambda_{0f}\ N_K^* \ .$$

In developing the expression for the effective cross section σ, we write

$$(4) \qquad R_K = n_G\ \sigma\ \frac{J}{e}\ \Delta V$$

where n_G is the number density of nitrogen molecules in the ground state, J is the electron current density, and ΔV is the interaction volume. Therefore,

$$(5) \qquad \sigma = \frac{e\ R_K}{n_G\ J\ \Delta V} \ .$$

Since $J = I/A$ and $\Delta V = LA$,

$$(6) \qquad \sigma = \frac{e\ R_K}{n_G\ I\ L}$$

where L is the length of the interaction volume.

If we define n_K^* as the number density of molecules in the excited state K, and n as the total molecular number density, we obtain

$$(7) \qquad n = n_G + \sum_K n_K^* = n_G + n^* .$$

Substituting into (6) we find

$$\frac{\sigma\ n_G\ I\ L}{e} = \lambda\ n^*\ \Delta V\ .$$

Substituting for n^* and rearranging leads to

$$\frac{n_G}{n} = \frac{e\lambda\Delta V}{\sigma IL + e\lambda\Delta V} \simeq 1$$

for typical operating values of the various parameters. Therefore, we may substitute n for n_G in (6).

$$R_K = \frac{n\ I\ L\ \sigma}{e} = N_K^* \sum_{f=0}^{n} \lambda_{0f}$$

$$= \lambda_{00}\ N_K^* \left[\sum_{f=0}^{n} \frac{\lambda_{0f}}{\lambda_{00}}\right]$$

or,

$$(8) \qquad \sigma = \frac{e}{n\ I\ L}\ \lambda_{00}\ N_K^* \left[\sum_{f=0}^{n} \frac{\lambda_{0f}}{\lambda_{00}}\right] .$$

The count rate of radiative transitions downward observed by the detection system is

$$C = \frac{d\Omega}{4\pi} \varepsilon \lambda_{00} N_K^*$$

where $d\Omega$ is the solid angle subtended by the detector and ε is the efficiency of the combined detection and counting system.

Upon substituting this and the ideal gas approximation into (8), we find our working equation for obtaining the excitation cross sections,

$$\sigma = \frac{4\pi k e}{d\Omega \varepsilon L} \left[\sum_{f=0}^{n} \frac{\lambda_{0f}}{\lambda_{00}} \right] \left[\frac{TC}{IP} \right] . \tag{9}$$

B. NUMERICAL VALUES

The following numerical values were measured or obtained:

$d\Omega = 0.023 \pm .001$ steradians

$L = 0.318 \pm .001$ cm

$k =$ Boltzman's constant

$\varepsilon \doteq (0.239 \pm .012)\%$

$e =$ electronic charge.

The λ_{0f} values were obtained from Nicolls [Ref. 3]

$\lambda_{00} = 1.11 \times 10^7 \text{ sec}^{-1}$

$\lambda_{01} = 7.27 \times 10^6 \text{ sec}^{-1}$

$\lambda_{02} = 2.83 \times 10^6 \text{ sec}^{-1}$

$\lambda_{03} = 9.33 \times 10^5 \text{ sec}^{-1}$

$\lambda_{04} = 2.54 \times 10^5 \text{ sec}^{-1}$

$\lambda_{05} = 5.11 \times 10^4 \text{ sec}^{-1}$.

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With these numerical values, equation (9) becomes in the working form

$$\sigma = (2.549 \pm .002) \times 10^{-32} \left[\frac{TC}{IP} \right] cm^2 \ .$$

Here T is temperature in degrees Kelvin, C is counts per second, I is the current in amperes, and P is the pressure in torr.

III. EXPERIMENTAL APPARATUS

The experimental apparatus consists of five major subsystems, each to be described later. They are: (1) the interaction chamber, (2) the vacuum system, (3) the electron beam generation system, (4) the optical system, and (5) the detection and counting system. Figure 1 is a diagramatic representation of the entire system.

A. INTERACTION CHAMBER

The electron-nitrogen collision events occurred in the interaction chamber (IAC) which was contained within a large aluminum enclosure. All connections from the electrical devices, the optical system, and the pressure measuring devices were passed through this enclosure to the IAC. The IAC (Fig. 2) itself was composed of two concentric chambers with the inner chamber electrically insulated from the outer chamber. The concentric chamber design was necessary to shield the interaction volume from an excessive number of stray electrons originating from the electron generator [Ref. 1]. The ends of both chambers nearest to the electron source were capped by a grounded flat circular plate with a one-eighth inch diameter hole in the center to admit the electron beam. The inner chamber also acted as a Faraday cup so that the actual beam interacting with the nitrogen molecules was measured. The grounded plate served a twofold purpose: it (1) allowed rough electronic alignment of the

electron beam, and (2) allowed a measure of the gross electron output of the electron generator system. The current required in the cross section equation was that obtained from the Faraday cup measurements.

The nitrogen gas was leaked into the IAC via a variable leak valve which allowed a constant density of nitrogen to be maintained with acceptable accuracy throughout the measuring process. This was insured by measuring the pressure with a Baratron barometer referenced to another vacuum system maintained below 10^{-6} torr pressure. The Baratron was capable of measuring pressure differences down to 10^{-5} torr. Thus when maintaining nitrogen at a pressure of 2×10^{-3} torr, the Baratron measured essentially the pressure needed for the cross section equation, since the reference vacuum may be assumed to be zero. Although ion guages were present for equipment and system evaluation, they were not utilized for pressure measurements during data taking because they acted as a source of unwanted light.

B. VACUUM SYSTEM

The vacuum was maintained by two six-inch diffusion pumps, rated at 1800 liters per second and 3000 liters per second respectively, discharging to a single fifteen-cubic-feet-per-minute two-stage mechanical pump. A liquid nitrogen cold trap was connected to each diffusion pump. The smaller diffusion pump was operated only during measurements to insure a controlled flow of nitrogen and to maintain the desired pressure in the IAC.

[illegible]

C. ELECTRON BEAM GENERATION SYSTEM

The electron beam was generated by a modified commercial television picture tube (RCA 7 JP-4). The tube was modified, as shown in Figure 3, by grounding lenses 2, 3, 4, and 5, as well as the end deflection plates. The indirectly heated cathode was also removed and replaced with a directly heated V-shaped 0.010-inch diameter tungsten wire cathode (filament), which could be removed for optical alignment of the beam generating device with the IAC.

The alignment required the lining up of the electron beam generating filament and the interaction chamber so that the electron beam would be guided through the lenses of the system and the grounded face plate of the IAC. Preliminary alignment was accomplished optically with a small HeNe laser directed along the path of the electrons. The filament was then aligned by adjusting the point of the V in the center of the extractor lens (lens 1). This alignment could be accomplished only when the system was open to atmospheric pressure and therefore with a cold filament, since access to the inner portions of the system was necessary.

To generate electrons, the cathode was heated with a small AC voltage and maintained at a negative bias. With the IAC at ground potential, the electrons were accelerated toward the face plate opening with an energy equal to the acceleration potential, or negative bias. With a small DC voltage applied to the deflection plates, compensation could be attempted for small misalignments of the beam. Some

problems were encountered in making these compensations, in that the response to the corrections of the deflection plates was inconsistent. This possibly was caused by filament droop resulting from heating the filament to extract the electrons. Since, as explained previously, the filament could be aligned only when cold, there was no way to correct any possible misalignment of the heated filament. This resulted in possible reduced currents at the Faraday cup.

D. OPTICAL SYSTEM

The optical system, shown in figure 4, collected photons emitted from the electron-nitrogen collision events at right angles from the incoming electron beam (see again figure 1 of the entire system) and focused them on the face of the photomultiplier tube for counting. The system was contained in a brass tube which was isolated from the IAC by a quartz window, thus allowing for removal of the optical system without affecting the vacuum of the entire system.

The optical system consisted of two positive lenses, a narrow band filter, and an aperture and field stop. The lens closest to the IAC (lens 1) was mounted so that its focal point was at the center of the electron beam and would pass parallel light through the filter to the other lens (lens 2). Lens 2 was mounted in the optical tube so that its focal point was at the face of the photomultiplier tube. The narrow band filter, which had a nominal transmission wavelength of 340 nm, was located between the lenses and

tilted 20° to the direction of the passing light so as to bring its peak transmission closer to the 337.13 nm wavelength which was to be measured. The circular aperture, immediately before lens 1 was 22.22 mm in diameter.

The unit magnification optics focused the incident light on a slit, 22.22 mm by 3.18 mm, in contact with the face of the photomultiplier tube. The long axis of the slit was oriented at right angles to the axis of the electron beam. The interaction volume was then defined by the width of the slit (3.18 mm) and the electron beam cross section. In the cross section equation; L is then fixed at 3.18 mm, and the solid angle ($d\Omega$) is determined by the aperture diameter and the focal length of lens 1 (13.06 cm).

E. DETECTION AND COUNTING SYSTEM

The photons collected by the optics were focused on and detected by an RCA 6810A photomultiplier tube. In order that the photomultiplier tube's dark count be maintained at its lowest possible level, the tube was cooled by liquid nitrogen boil-off passed through a copper jacket surrounding the tube. The liquid nitrogen boil-off was channeled through a Union Carbide TC-1 Liquid Nitrogen Controller which insured that only gaseous nitrogen passed to the tube's insulating jacket, so that its linearity and stability were not destroyed by the extreme cold, and also regulated the temperature of the gas in the jacket to maintain consistency and eliminate temperature gradients across the tube. This was accomplished by a thermistor located at the insulating jacket's exhaust.

The temperature of the tube could be measured with the aid of another thermistor placed between the jacket and the tube.

To prevent the possible frosting or fogging of the photomultiplier tube face or lens 2 of the optical system because of the extreme cold, a defogging chamber was located between the photomultiplier tube and the optics tube. A portable vacuum pump was used to evacuate the defogging chamber and then the chamber was sealed with a small vacuum valve and continually measured by a vacuum gauge.

The signal from the photomultiplier tube was then passed through a preamplifier, amplifier, and pulse height analyzer to a scaler-timer. This equipment was set at the optimum signal-to-noise ratio setting determined with a model 590 EG and G calibrated lamp system. The efficiency of the optical and detection systems was (.229 ± .012)% [Ref. 4]. This then was the efficiency (ε) required in the cross section equation.

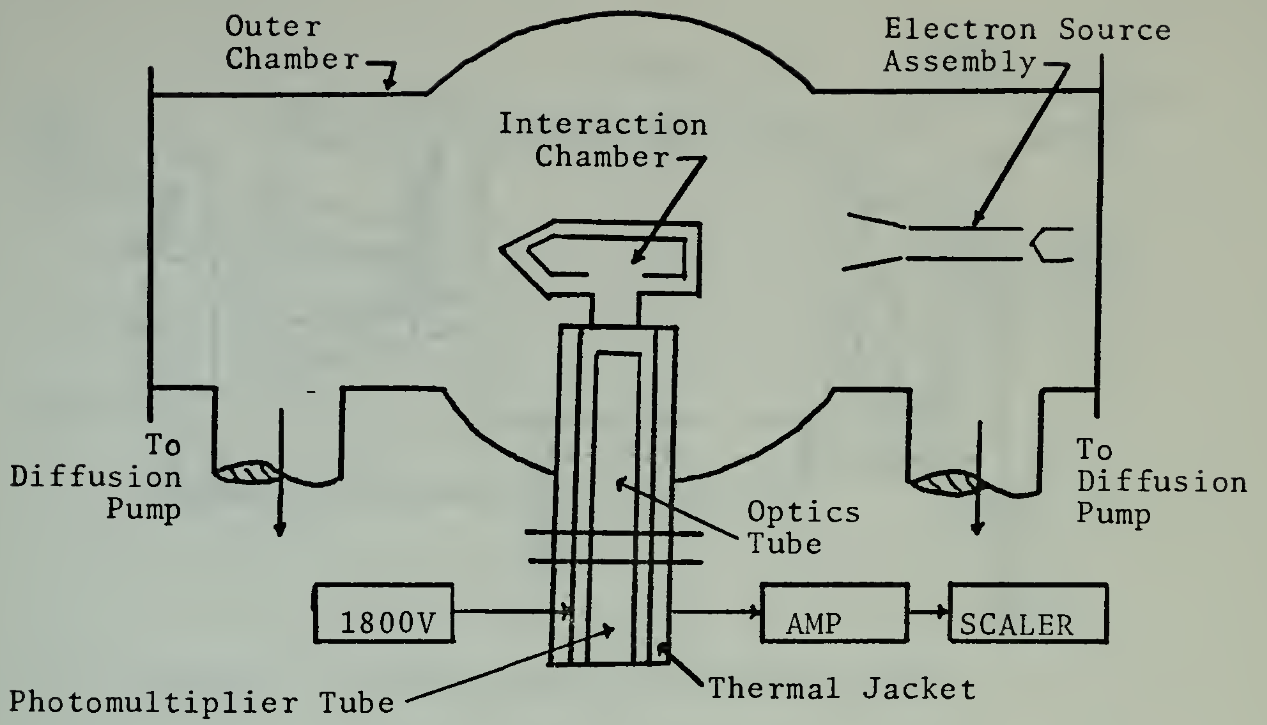

Figure 1. Experimental Apparatus

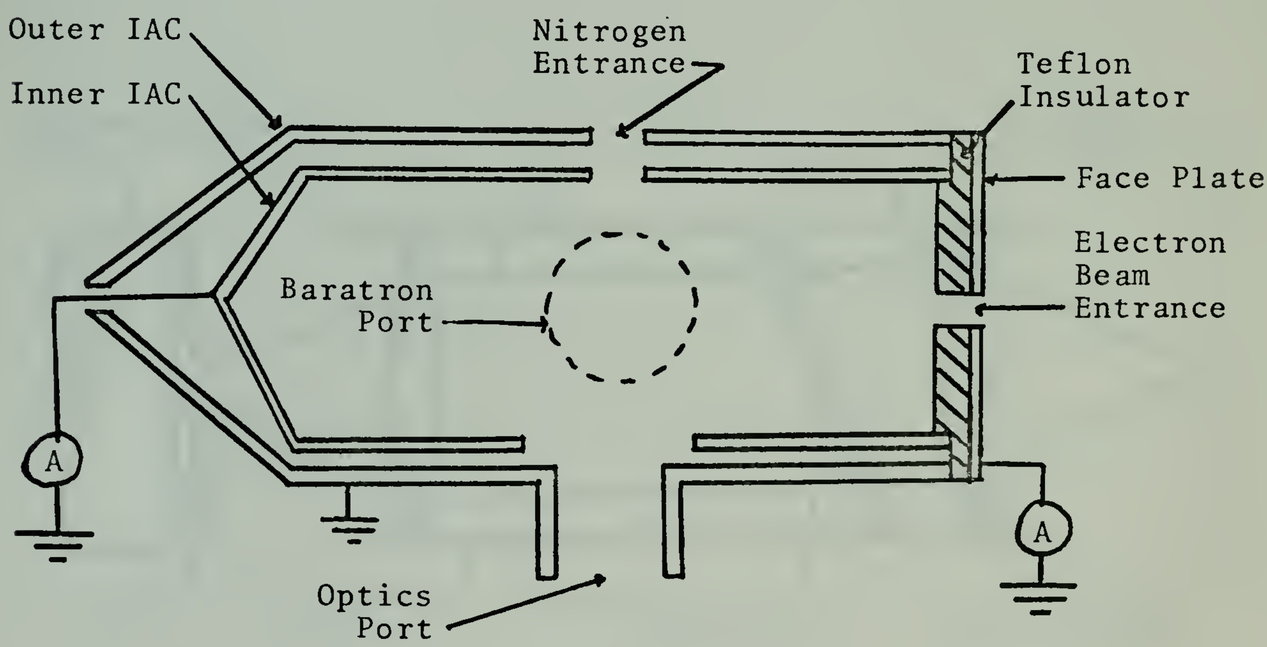

Figure 2. Interaction Chamber

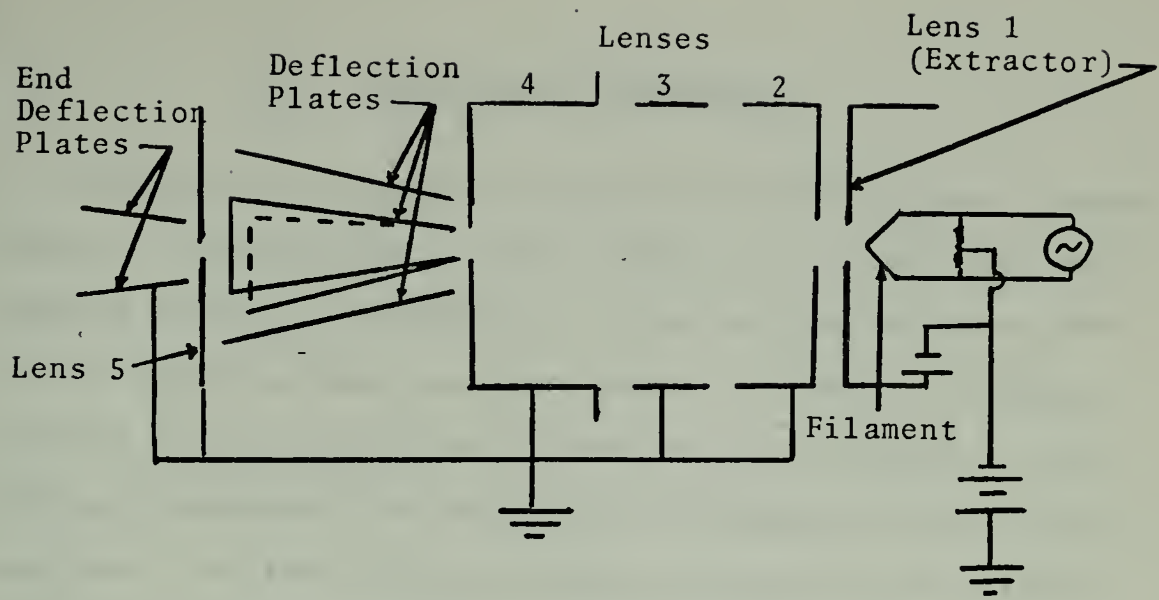

Figure 3. Electron Beam Generation System

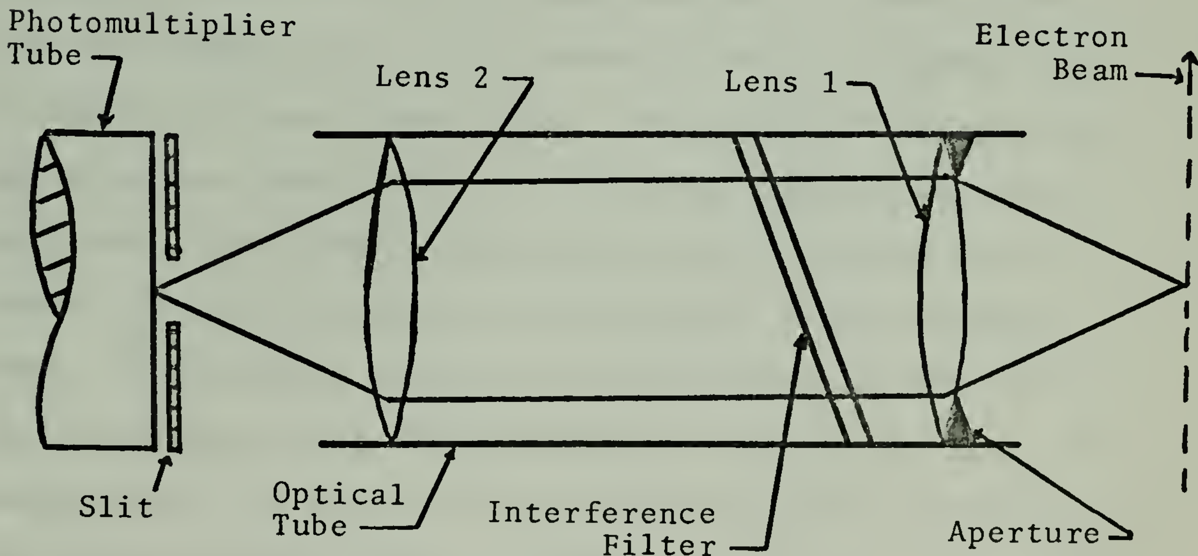

Figure 4. Optical System

IV. EXPERIMENTAL PROCEDURES

The electron energies selected for the experiment ranged from 80 to 900 electron volts. Below 80 eV there was insufficient current to determine a cross section and above 900 eV the electron beam generator showed evidence of arcing, causing great fluctuations in the current produced in the IAC and precluding any reasonable measurement of the cross sections. At the electron energies selected, the current and gas pressure in the IAC were measured, and the temperature in close proximity to the chamber was measured. Since all parts of the system had to be operating for several hours in order to stabilize the system prior to data measurement, and there were no heat sources or sinks in the IAC, the temperature in the chamber could be assumed to approximate the room temperature. Ten counts of ten second duration were then taken at each energy setting for both dark and light current during each run. This was done to reduce any error induced by fluctuations in the electron beam. The filament voltage was left on during dark counts to insure that light from the filament was not biasing the measurements. A series of data runs were made to eliminate any extraordinary effects or malfunctions.

As explained in the theory portion of Section II, the collisional deexcitation rate could be considered negligible when the gas pressure was less than 3×10^{-3} torr. During

[illegible]

the entire series of data collection runs, a gas pressure not higher than 2.1 x 10^{-3} torr was used. At the energies selected with gas pressures in the vicinity of 2 x 10^{-3} torr, the current in the beam was consistently in the 10^{-7} ampere range. This was sufficient to produce adequate counts to make good measurements of the cross sections.

V. RESULTS

The effective cross section measurements ranged from a maximum value of $(1.72 \pm .22) \times 10^{-17}$ cm^2 at 250 eV to $(9.47 \pm 2.53) \times 10^{-18}$ cm^2 at 80 eV. The results were summarized in figure 5.

Included in the total error estimates of the individual cross section per run were: (1) inaccuracies in reading measured temperature, pressure and electron beam current, (2) the error in measuring the efficiency, which was determined by the error in lamp calibration as reported by the manufacturer and the statistical standard deviation of the counts taken during the calibration process, and (3) the statistical standard deviation of the counts taken during the data runs. The cross sections at each energy were assumed to obey a normal distribution and the errors shown in figure 5 were calculated accordingly. The large standard deviations both at the high and low energy ends reflect the aforementioned difficulties at these energies.

The cross section at 125 eV is consistently higher than expected and bears further investigation.

Poole [Ref. 4] measured a maximum cross section of 1.4×10^{-17} cm^2 at 150 eV. His results are also shown in figure 5. His results are expected to be less accurate than ours since he completed only one data run and operated at a pressure much greater than that previously determined as an upper limit for allowing the collisional deexcitation rate

Q to be considered negligible. The effects of a collisional deexcitation rate would depress the cross section values, which could account for the large differences between his values and ours. Poole also could not make measurements below 150eV, which prevented him from positively identifying a peak cross section.

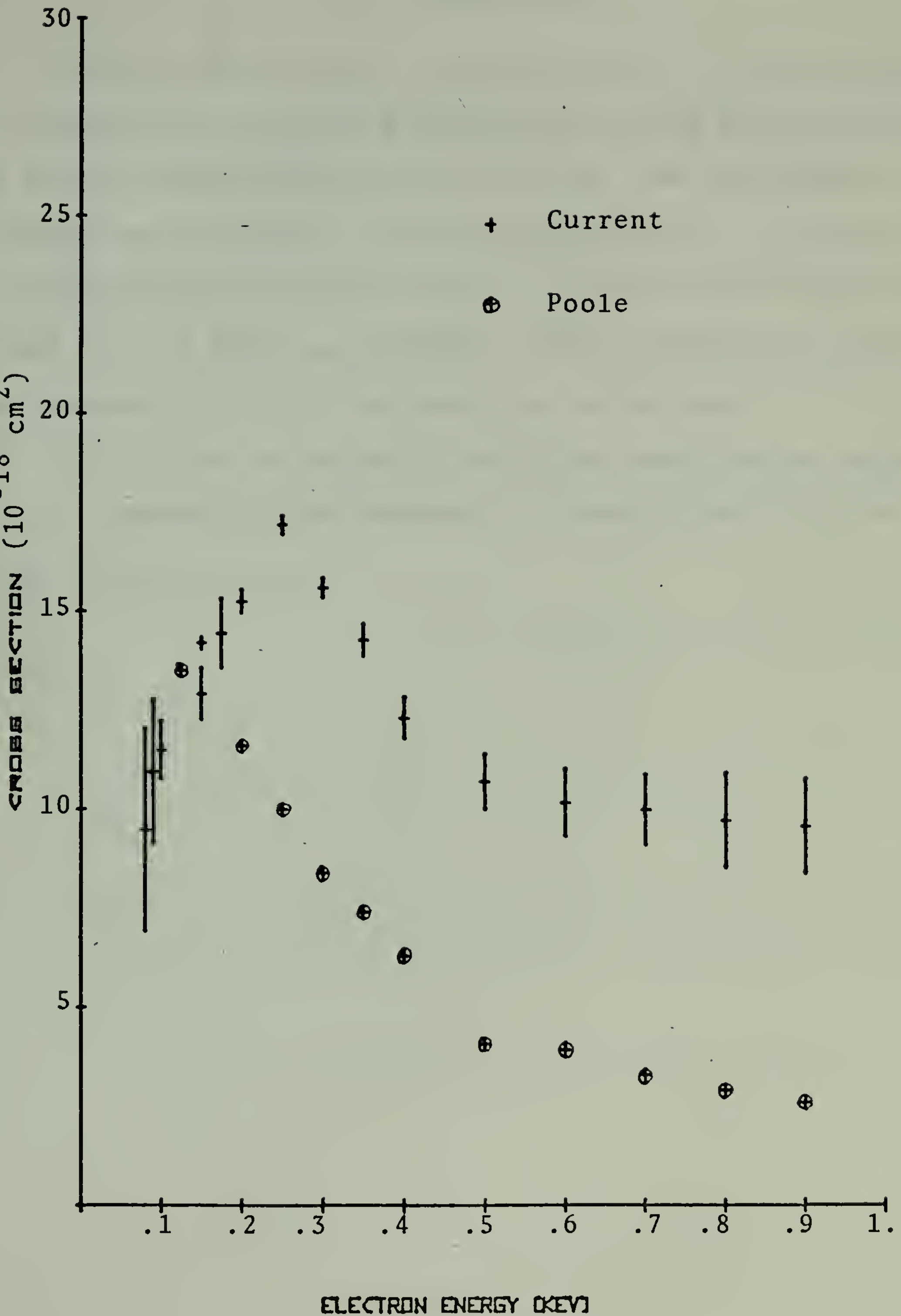
30
25
20
15
10
5
CROSS SECTION (10^{-18} cm^2)
Current
Poole
.1
.2
.3
.4
.5
.6
.7
.8
.9
1.
ELECTRON ENERGY (KEV)

Figure 5.

VI. CONCLUSIONS

Further measurements are precluded until the equipment is redesigned to allow a better controlled electron beam and a higher resolution detection system. We were unable to determine the reason for the measurements at 125 eV and recommend further measurements in this energy range be carried out. A more controllable electron beam would allow measurements at very low energies to be made.

The system is suitable, with the above rework accomplished, to conducting measurements of other gases, such as oxygen and nitrogen monoxide.

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